Review of 'A global tropospheric ozone climatology from trajectory-mapped ozone soundings', by G. Liu et al.

General Comments

This paper presents an interesting new observation-based tropospheric ozone climatology. Ozone sonde data are combined with backwards and forwards trajectories (up to 4 days) to construct ozone fields over a much wider area than just the sites of the sondes. The authors nicely demonstrate the utility of this method and the climatology itself. This is the main purpose of the paper, and it is well done. However, I take issue with some other parts of the paper.

Firstly, the authors should document how convection is handled in the trajectory modelling. This will be crucial in the tropics, but is not mentioned.

Secondly, the comparison with satellite data of total column ozone could be better presented by constructing ozone columns from the climatology, to allow a direct comparison with the data.

Thirdly – there are some odd statements about the seasonal cycle of ozone that need some rethinking.

Finally – and perhaps most importantly – there are sweeping and controversial conclusions drawn about ozone trends, that I suspect are not justified. The climatology shows little trend in ozone since 1970, which disagrees with some other studies, which use different sources of data. My suspicion is that this climatology contains quite large uncertainties associated with changes in ozone sonde measurement techniques, and that these are large enough to preclude making definitive statements about trends. The authors need to clarify this section and probably tone down their statements about trends, or alternatively bolster their findings to make them more believable.

If these points can be adequately addressed, I think this paper will make an interesting and very useful contribution to the literature on tropospheric ozone.

Specific Comments

p11476 l15 'intensives' is shorthand slang. I think you mean 'intensive campaigns'.

p11476 l17 The acronym WOUDC should be defined at first use (not a few lines later).

p11477 l12 Smit et al (2007) is missing.

p11480 l9 You indicate that you can see differences between how emissions are 'lofted' in different regions, which raises the question of modes of vertical transport. In the description of the trajectory modelling, some mention should be made of how convection is handled. Convection is presumably largely sub-grid-scale for the meteorological reanalysis data used to calculate trajectories, and is thus not included. Yet convection, especially in the tropics, is a crucial mechanism for transporting and mixing species in the vertical, including ozone. Some trajectory modelling applies techniques to account for convection – do you? Even on short timescales of a few days, ignoring convection may have important consequences that should at least be mentioned.

p11481 l23 Neglect of deposition is another important caveat to note for near-surface results.

p11483 l26 Figure 6 shows all months, not 'selected'.

p11483 l28 Why don't you show a like-for-like comparison, rather than comparing modelled ozone at specific levels with a column ozone for the whole troposphere? It would be relatively simple to convert your climatology into tropospheric ozone columns, to allow a direct comparison with OMI/MLS. This would also facilitate a comparison with Chemistry-Climate Model results, e.g. Young et al (2013, Figure 6).

p11484 l15 You discuss the seasonal signature of ozone in the 'hemispheric average'. This is a rather crude average, and masks large differences in both latitude and longitude. Royal Society (2008) explored this within models (Box 5.2, p44) – e.g., in northern mid-latitudes, a clear spring peak is seen over remote regions - oceans, but a summer peak is seen over land – this discrimination would be lost in a hemispheric average. I suggest your observational climatology offers a great opportunity to generate an observation-based version of this sort of analysis (and not just at the surface).

p11484 l19- You relate the NH spring peak (previous comment) to the annual cycle in stratospheric ozone, suggesting this is consistent with current models 'that find stratospheric input to be similar in magnitude to net photochemical production'. I struggle to see the link here. The implication is that the spring peak in ozone is a 'global phenomenon' (when is spring in the tropics?) - as I state above, I think it is a bit more complex than that – and also that this spring peak is driven by the stratospheric cycle, with a lag. I think this is far from obvious and remains an open question. There are several contributing factors to the spring peak in ozone in the remote northern hemisphere mid-latitude troposphere – the stratosphere is one, but tropospheric photochemistry and the build-up of ozone precursors through the winter is clearly another (and there are several others). Just because models find that the magnitude of stratospheric input to be similar to the magnitude of net photochemical production does not imply a link. This relationship is simply a consequence of the fact that net chemical production must balance the net physical transport/removal of ozone from the troposphere. Models estimate that net stratosphere to troposphere transport is ~500 Tg(O_3)/yr, and that dry deposition of ozone at the surface is ~1000 Tg(O_3)/yr. By mass balance, it follows that net chemical production of ozone must be ~500 Tg(O_3)/yr. The fact that the stratospheric source and net chemical production are similar in magnitude tells us nothing about the factors driving the seasonality in tropospheric ozone.

p11484 l24 The text suggests Figure 8 is for 1-2km and Figure 9 is for 4-5km, but the figure captions suggest the opposite.

p11485 l6 'same data' – Figures 8 and 9 are based on your trajectory modelling of ozone sonde data, whereas the studies you refer to mainly use either data from surface stations or MOZAIC aircraft data. So I don't think these can be described as the 'same data'.

p11485 l6 'in no way contradict these results' – I disagree. Either there has been a trend in tropospheric ozone or there has not – or the data are inconclusive. The other studies you refer to find generally upwards trends in ozone, for at least part of the record (as you summarise). You find little trend over the last 30 years. So you should clarify what you are saying, and I think it either has to be: (i) the earlier studies were wrong, and there is no trend, or (ii) the data are too uncertain to be conclusive either way.

p11485 l10 ‘even lower relative to ECC sondes’ – please clarify exactly what you mean.

p11485 l14 Clarify what you mean by ‘ratio of ozone concentration’ – ratio of what to what? (I think you mean relative to the 2000s)

p11485 l15 Clarify ‘corrections’. This is the crux of the matter, I think. Does the uncertainty associated with these corrections preclude using the data to show significant trends? I suspect the answer to this question is yes. You must clarify this.

p11485 l20 ‘suggest the surprising view that globally, free tropospheric ozone has not changed very much in the past four decades’. To me, this statement does disagree with the findings of Parrish et al, Cooper et al., etc., so I think you are contradicting their results. I find it hard to square this statement with the statement on line 6 of this page.

p11486 l3 ‘reasonable agreement’ – Can you be more quantitative? Within 10%, 20%, 50%?

p11486 l12 ‘modest’, changes ‘largely instrumental in origin’ – these statements need considerable caveats (see previous comments).

p11486 l19 – comparison with TOC retrievals – as previously commented, please demonstrate this comparison more quantitatively.

p11507 Figure 10 caption – it is unclear what is meant by ‘a polynomial fit…’

Reference

Royal Society, Ground-level ozone in the 21st century: future trends, impacts and policy implications, 2008 (http://royalsociety.org/policy/publications/2008/ground-level-ozone/)